Quantifying exchange coupling in segregated granular materials

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The volume of a magnetic grain, together with its anisotropy, determines the probability of thermally activated reversal. Thus for grain volume distributions where the median volume is close to the superparamagnetic limit there will be a sub-set of grains which are either superparamagnetic on the time scale of a typical magnetic measurement (10s), or reverse due to magnetostatic fields from surrounding grains. We use this effect to probe exchange coupling in segregated granular materials, using CoCrPt-SiO_x granular recording media as model systems. As the film thickness is reduced below 10 nm the remanent magnetization of these films decreases, due to thermal activation and magnetostatic reversal. Varying film thickness and temperature allows us to thermally select a population of grains that contribute to the measurement. Exchange coupling is characterized by the angle dependence of remanent coercivity where we associate a breaking of symmetry from the Stoner-Wohlfarth model towards the Kondorsky model as a measure of the incoherency of reversal. Combining these models allows an estimate to be made of the volume fraction of grains that are exchange coupled and we find that, for well segregated CoCrPt-SiO_x media, approximately 8% of the magnetic volume undergoes some degree of exchange coupling.

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High anisotropy, segregated, granular CoCrPt-SiO_x thin films are key materials in perpendicular magnetic recording [1]. When incorporated into an exchange coupled composite medium, by layering with a lower anisotropy magnetic material, the maximum recording density can be increased towards 1 Tbit/in^2, with no loss in writeability [2,3]. More generally, nano/micro-scale granular materials are also highly applicable as permanent magnet materials [4-6]. It is important to understand how grains interact in these granular materials, and the effect these interactions have on magnetic reversal, especially when considering a more complex composite material where simple models do not adequately describe the switching process. The origins of incoherency in granular CoCrPt systems have been studied previously [7], and the effect of intergranular exchange coupling has been studied by creating a model multilayer system [8,9] and through measurements of real CoCrPt granular media [10-13]. The contribution of inhomogeneous microstructure to intergranular coupling has also been studied [14].

In this paper, we report the switching behaviour of granular materials with a grain size distribution in thermal regimes above and below room temperature. The volume of the individual grains is controlled by tuning the thickness of the thin film. An example grain volume distribution is shown in figure 1. This allows us to distinguish the effects of thermal activation from inter-granular exchange coupling, confirming the methodology that we have developed previously [12,13] across a range of film thicknesses and temperatures. Due to reduced magnetocrystalline anisotropy energy K_uV (K_u is the magnetocrystalline anisotropy and V is the volume) in thin films, we are able to thermally select sub-sections of the grain population for magnetic remanence measurements. In these measurements, only grains that switch during application of a reverse field are measured i.e. the grains that contribute to the remanent state. In weakly exchange coupled granular systems, grains will contribute to the remanence if their anisotropy energy is sufficient or if they are exchange coupled to neighbouring grains. This exchange coupling leads to symmetry breaking in magnetic reversal caused by incoherency, and this is explored as an intrinsic property close to the limit of granular thermal stability.

The granular materials used in this study were heterogeneous CoCrPt-SiO_x granular media intended for perpendicular magnetic recording. The samples were deposited via dc magnetron sputtering onto 65mm diameter, 0.6mm thick glass substrates. Initially a 30nm thick CoFe-based soft underlayer (SUL) structure was deposited onto the substrate, followed by a Ru seed layer, and then a CoCrPt-SiO_x granular segregated recording layer of thicknesses from 4nm to 16nm. Finally, the media were capped with a protective carbon overlayer and a lubricant.

DC demagnetizing remanence curves were measured using a Microsense DMS model 10 vector vibrating sample magnetometer (VSM), with temperature control and a maximum field of 20 kOe. Due to the presence of the SUL, a measurement technique is required that can separate the measured magnetic moment due to the SUL from that of the granular recording layer, which is of interest [15]. This involves application of an in-plane pinning field during the measurement of remanent moment perpendicular to the film plane. Applying the field at an arbitrary angle allows a series of remanence curves to be measured.

Remanence curves were measured as a function of applied field angle relative to the easy axis of the material (perpendicular to the plane), for CoCrPt-SiO_x films with thicknesses ranging from 4 nm to 11 nm. Example remanence curves measured for films of 6 and 11nm at two angles (0º and 45º), are shown in figure 2. When the film thickness is reduced to 6 nm the material is no longer fully remanent due to thermal activation and magnetostatic reversal of the smallest grains in the grain size distribution. The remanent magnetization (saturation point in the remanence curve) is plotted in figure 3 as a function of film thickness. These data clearly show that for films 10nm thick and below a proportion of the grain population is no longer remanent, as illustrated in figure 1. This indicates that we are thermally selecting a population of grains based on the grain volume distribution, the temperature and the coupling (exchange or magnetostatic) between grains.

The remanent coercivity H_{cr} at room temperature was extracted from the raw remanence data and then plotted as a function of angle between 0º and 60º for each film thickness, as shown in figure 4. The remanence curves are expected to exhibit behaviour between the two limits of the Stoner-Wohlfarth model [16] (coherent rotation for isolated particles) and the Kondorsky model [17] (domain wall motion in highly exchange coupled systems). There is a dispersion of easy axes in the granular material [15], which has a Gaussian form and is characterized by σ = 3°. This results in the measured remanence curve being the sum of curves for individual grains with a mosaic spread of easy axes of anisotropy. This results in a change in the functional form of the reversal curve, with a minimum switching field of $0.58H_{cr}(0º)$ (Compared to $0.5H_{cr}(0º)$ for the unmodified Stoner-Wohlfarth model). CoCrPt-SiO_x. thin films of 11nm thickness have been shown in previous work, within the resolution of the technique, to exhibit switching behaviour that demonstrates coherent, exchange-decoupled switching of individual grains [12]. We therefore use this thickness as a reference to explore the evolution of incoherency in reversal as the thickness is reduced.

Figure 4 shows the evolution of the switching behaviour as the thickness of the films is varied between 6 nm and 11 nm. For a small decrease in the thickness of the film from the optimal (fully remanent) case of 11nm the switching behaviour remains unchanged. As the thickness of the layer is decreased below 10nm, the switching field minimum becomes shallower, and occurs at a smaller angle, reaching a minimum at 41° for the thinnest film (6nm) as shown in figure 5(a). This can be attributed to an increased degree of incoherent switching in the material relative to the total, e.g. the ratio $M_{inc}/(M_{inc}+M_{coh})$ has increased, where M_{inc} and M_{coh} are the total magnetizations that switch incoherently and coherently, respectively. Our previous results show that thermal activation only changes the magnitude of the normalized minimum switching field, and not the angle at which this minimum occurs [12]. The reduction in remanent magnetization is therefore not simply due to the shift in the mean anisotropy energy to lower values – if this were the case, all the remanent grains would switch coherently, and this is not supported by the data in figures 4 and 5(a). A film with thickness 4nm was also measured (figure 3 insert), and showed reversal behaviour with significant incoherency, the curve more closely resembling the Kondorsky model of switching ($H_{cr} \propto \cos(\theta)^{-1}$).

The change in angle dependent switching in sub-10nm thick films cannot be due to reduced coherency within the grains. Individual grains have lateral dimensions roughly equal to the exchange length in CoCrPt, which may be estimated using the expression $l_{ex} = \sqrt{A/K_u}$, where A is the exchange constant and K_u is the uniaxial anisotropy of the grain, including both magnetocrystalline and shape terms. Inserting values of $A = 1\times10^{-6}$erg/cm and $K_u = 5\times10^{6}$erg/cm^3 yields $l_{ex} \sim 5$nm. Therefore individual grains might be expected to switch coherently, were they in complete isolation. However, coupling with adjacent grains provides a mechanism for the observed incoherency. Inter-granular coupling is normally assumed to be negligible, as evidenced by the switching curve for the 11nm film, which exhibits a minimum switching angle of 45º, consistent with the Stoner-Wohlfarth model of coherent switching (isolated grains). However, due to thermal activation or magnetostatic interactions, there will be a population of grains where either the grain volume is too small for the grains to be remanent or the grains will orient in the opposite direction. These grains would either not individually participate in the remanence experiment (thermal activation) or reduce the net remanent magnetization (magnetostatic interaction). This can be seen from figure 3, where for film thicknesses below 10nm the remanent magnetization begins to decrease. In order for these grains to participate or align with neighbouring grains, we hypothesize that they must be coupled via inter-granular exchange coupling, thus enhancing the effective volume of the grain allowing them to be thermally or magnetostatically stable under the experimental conditions employed (T = 292 K, t = 10 seconds). These grain ensembles can reverse incoherently, due to increased lateral dimensions and coupling effects. Another possible explanation could be that the first few layers of magnetic material grow with poor granularity, as has been seen previously, and this causes incoherent reversal. However, from figure 4 it can be seen that once the film thickness reaches 10nm, coherent reversal is measured as this is the dominant contribution. This would not be possible if a percentage of the thickness were poorly segregated.

Figure 5(b) shows the minimum switching field angles for the remanence curves shown in figure 4, plotted as a function of squareness (M_r/M_s). Below a threshold value of approximately 0.8, the minimum angle begins to reduce. This indicates that the smallest 20% of the grains become thermally unstable or misaligned and cannot be stabilized in the field direction by exchange coupling to neighbouring grains. Beyond this point increased incoherency is observed, due to the enhanced contribution of coupled grains to the measured remanence.

Measurements of angle dependence of remanence for the 6 nm thick film, over a range of temperatures, are plotted in figure 6. More uncertainty is visible in the curves at higher temperatures due to increased error in the extracted values of H_{cr}. This arises because of a reduced signal-to-noise ratio in the raw data, as the remanent magnetization is reduced due to thermal activation and magnetostatic reversal. In contrast to previous temperature dependent measurements on thicker films (11nm and 16nm) [12], the minimum switching field angle varies as a function of temperature. This suggests that by increasing the temperature we are sampling a larger proportion of coupled grains, resulting in more incoherent reversal. Temperature therefore provides an additional method of probing inter-granular coupling provided that the structural properties of the material remain stable.

Figure 7 shows the variation in minimum switching angle plotted as a function of M_r/M_s for the data in figure 6. An approximately linear relationship is observed. Extrapolating a linear fit to the data to zero remanence allows us to estimate the maximum reduction in minimum switching angle for the 6nm thick film (39±2 deg). Using a model that comprises a weighted average of two switching regimes (Kondorsky-like for coupled grains, Stoner-Wohlfarth coherent switching for un-coupled grains),

$$\frac{H_{cr}(\theta)}{H_{cr}(0)} = \frac{x}{\cos(\theta)} + (1-x)H_{S-W}(\theta), \qquad (1)$$

where x is the fraction of inter-granularly coupled material, and $H_{S\text{-}W}(\theta)$ is the Stoner-Wohlfarth switching field incorporating a 3° dispersion of easy axes. This function is plotted in figure 8 for six values of x. The validity of this zero temperature approach has been established in previous work, through micromagnetic simulation and experiment [12,13]. We demonstrated that thermal activation changes only the depth of the minimum, not the angle at which it occurs. We therefore compare only the minimum angle and exclude the effects of thermal activation and the temperature dependence of material constants. The closest match to the experimental minimum switching angle is for x=8±3%, corresponding to an estimated 8% of remanent magnetic material being coupled. This is the first estimate that has been made of the volume of magnetization that is coupled in granular media. The results demonstrate that with a properly controlled set of samples, an estimate of the absolute proportion of coupled grains may be obtained. This opens the possibility of both comparing different materials and process conditions and comparing experimental data to simulation.

In conclusion, we have performed a detailed study of angular remanence as a function of CoCrPt-Ox. film thickness and temperature. By reducing the mean anisotropy energy we are able to thermally select a population of grains for which the effects of inter-granular exchange coupling are detectable through angular remanence measurements. We have shown that from these measurements the volume of magnetization that is exchange coupled in granular magnetic materials can be estimated. The incoherency observed in reversal is due to the exchange coupling of smaller grains that would otherwise not participate in the experiment individually due to thermal activation or switching as a result of magnetostatic fields. Our method identifies incoherency by symmetry breaking from the Stoner-Wohlfarth (S-W) model towards a hybrid S-W/Kondorsky model. Comparison of the minimum switching field angle values from experiment to the hybrid S-W/Kondorsky model allows an estimate to be made of the percentage of coupled grains (8±3%) in our samples. The estimate is obtained by extrapolating to find the minimum switching field angle at zero remanence as this point represents the maximum sensitivity to coupled grains. The technique described here can be applied more generally to explore inter-granular exchange coupling and coherency of reversal in granular magnets that comprise nominally decoupled grains at optimal thicknesses and temperatures.

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References

[1] Piramanayagam S 2007 *J. Appl. Phys.* **102** 011301

[2] Dittrich R, Schrefl T, Kirschner M, Suess D, Hrkac G, Dorfbauer F, Ertl O and Fidler J 2005 *IEEE Trans. Magn.* **41** 3592

[3] Victora R H and Shen X 2005 *IEEE Trans. Magn.* **41** 2828

[4] Shinba Y, Konno T J, Ishikawa K, Hiraga K and Sagawa M 2005 *J. Appl. Phys.* **97** 053504

[5] Matsuura M, Sugimoto S, Goto R and Tezuka N 2009 *J. Appl. Phys.* **105** 07A741

[6] Hrkac G, Woodcock T G, Freeman C, Goncharov A, Dean J, Schrefl T and Gutfleisch O 2010 *Appl. Phys. Lett.* **97** 232511

[7] Jung H S, Kuo M, Malhotra S S and Bertero G 2007 *Appl. Phys. Lett.* **91** 212502

[8] Sokalski V, Laughlin D E and Zhu J G 2009 *Appl. Phys. Lett.* **95** 102507

[9] Sokalski V M, Zhu J G and Laughlin D E 2010 *IEEE Trans. Magn.* **46** 2260

[10] Berger A 2012 *Physica B: Cond. Mat.* **407** 1322

[11] Brandt R, Tibus S, Springer F, Fassbender J, Rohrmann H, Albrecht M and Schmidt H 2012 *J. Appl. Phys.* **112** 033918

[12] Morrison C, Saharan L, Hrkac G, Schrefl T, Ikeda Y, Takano K, Miles J J and Thomson T 2011 *Appl. Phys. Lett.* **99** 132507

[13] Saharan L, Morrison C, Miles J J, Thomson T, Schrefl T and Hrkac G 2011 *J. Appl. Phys.* **110** 103906

[14] Tham K K, Saito S, Hasegawa D, Itagaki N, Hinata S, Ishibashi S and Takahashi M 2012 *J. Appl. Phys.* **112** 093917

[15] Thomson T, Lengsfield B, Do H and Terris B D 2008 *J. Appl. Phys.* **103** 07F548

[16] Stoner E C and Wohlfarth E P 1948 *Phil. Trans. R. Soc. Lond. A* **240** 599

[17] E. Kondorsky 1940 *J. Phys. Moscow* **2** 161

Figures:

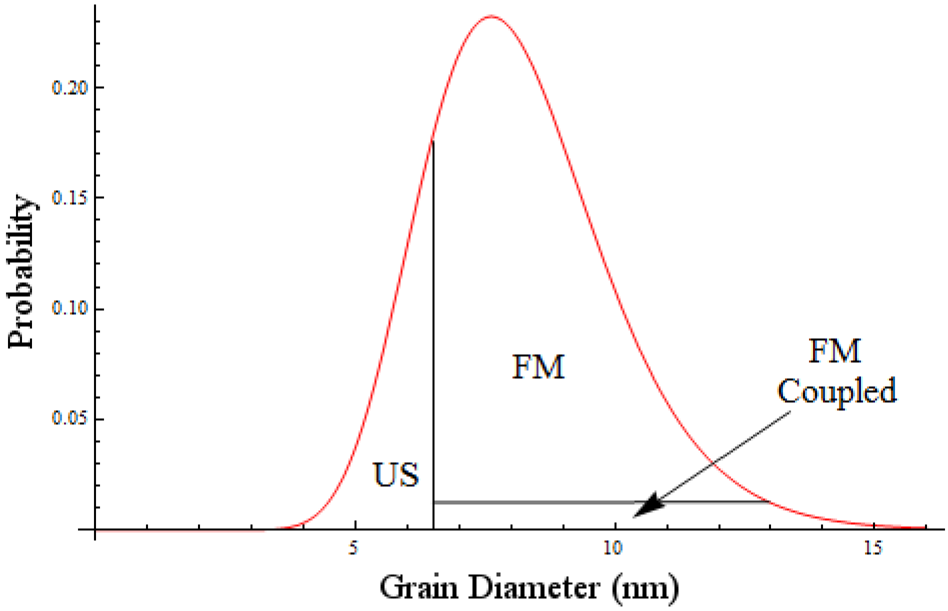

Figure 1: Illustration of unstable (US), ferromagnetic regions that exhibit intergranular exchange coupling (FM Coupled) and ferromagnetic (FM) regions in the grain diameter distribution for a 6nm thick film.

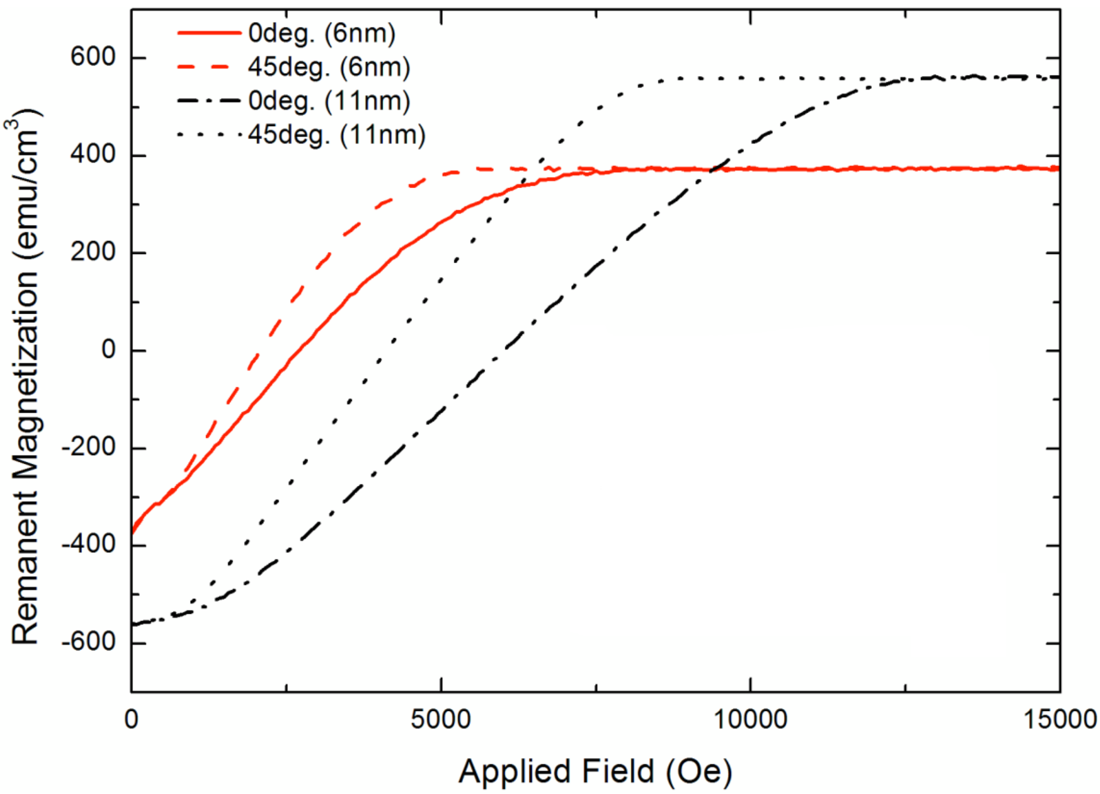

Figure 2: Example remanence measurements for CoCrPt-Ox. films of 6 and 11nm thickness, at angles of 0 and 45° relative to the perpendicular anisotropy axis, measured by vector vibrating sample magnetometry. The 11nm film is fully remanent at room temperature; the 6nm film is no longer fully remanent due to thermal excitation and/or magnetostatic reversal of the smallest grains in the film.

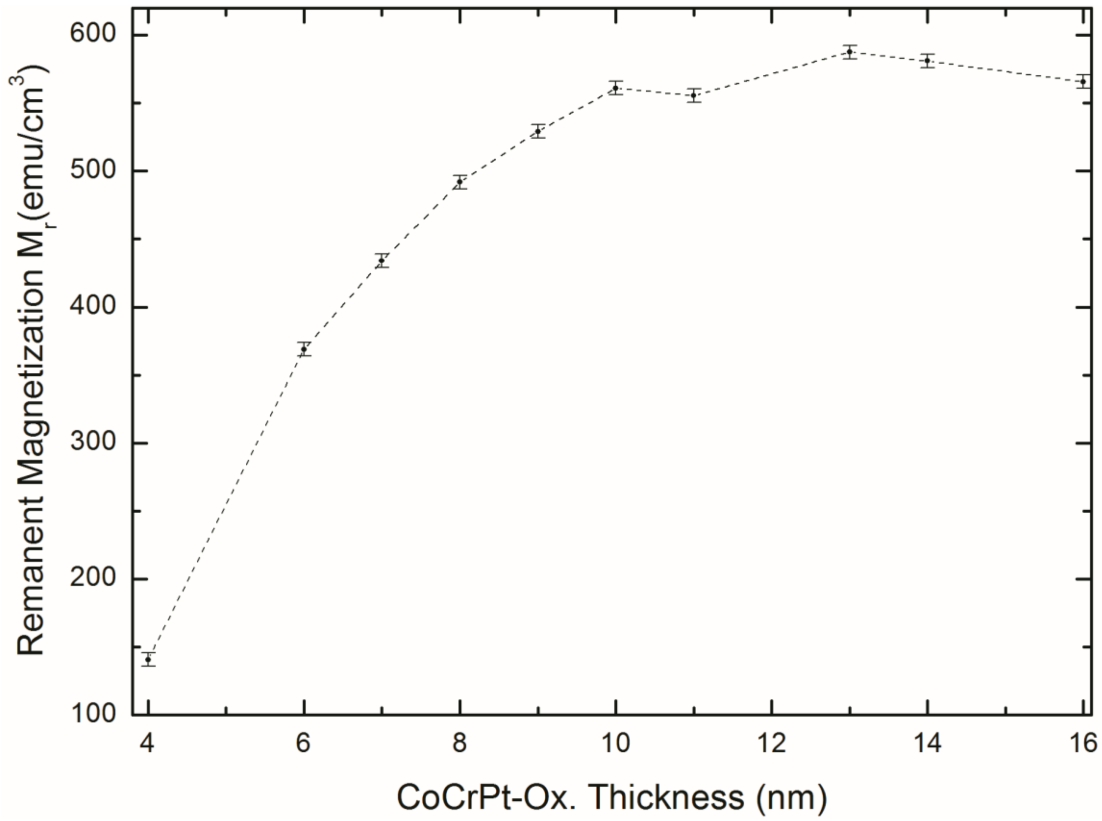

Figure 3: Remanent magnetization M_r plotted as a function of CoCrPt-Ox. recording layer thickness. Below 10nm thickness M_r starts to reduce, indicating a fraction of the grain size distribution becomes unmeasurable due to thermal activation or magnetostatic reversal. The dashed line is a guide to the eye.

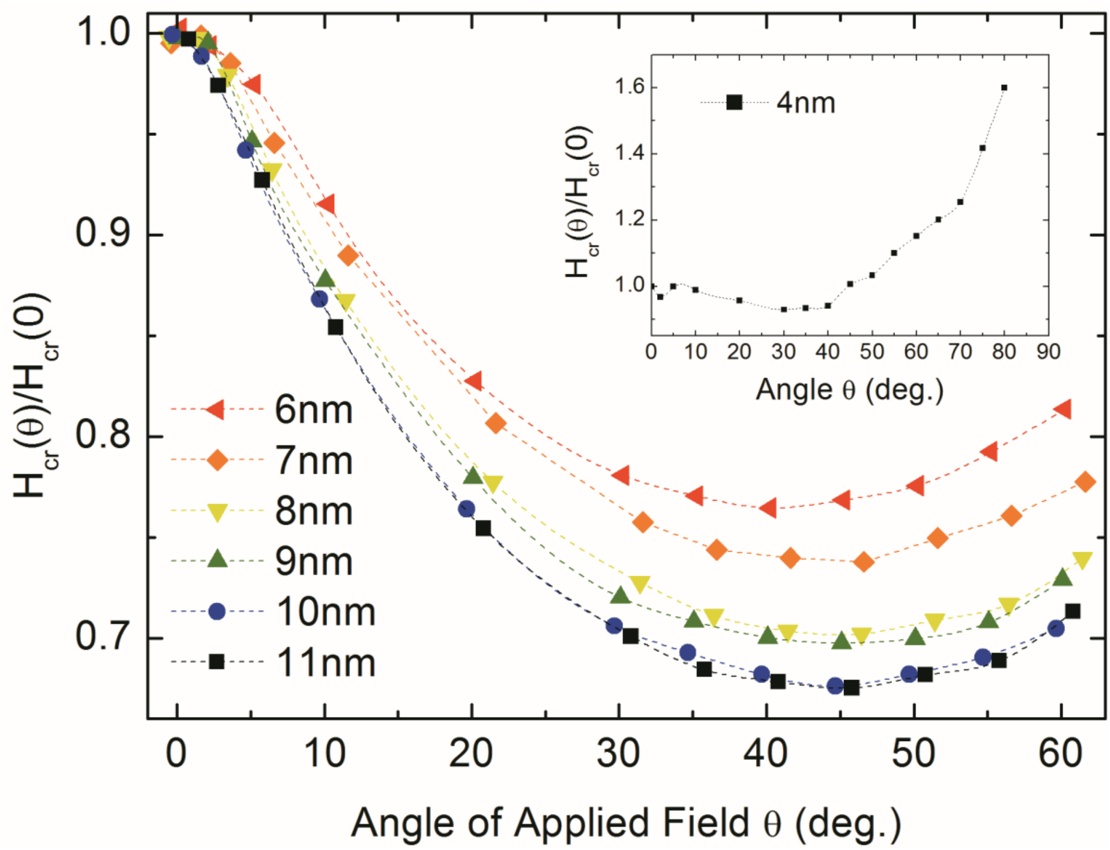

Figure 4: Remanent coercivity H_{cr} as a function of film thickness. A clear evolution in the function form of the curves indicates reduced coherency of switching as film thickness is reduced. Inset: Data for a 4nm thick film showing highly incoherent reversal. Dashed lines are a guide to the eye.

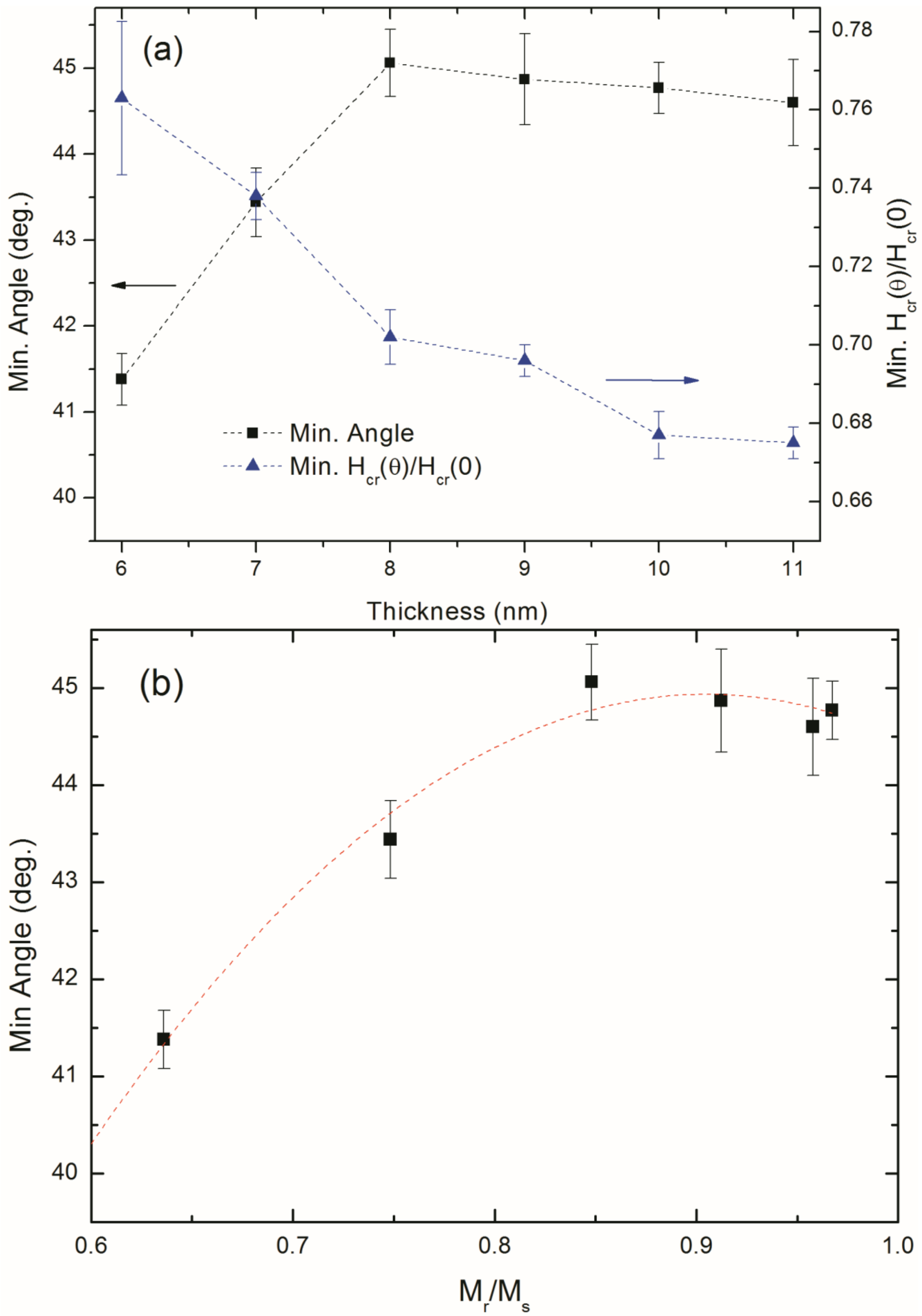

Figure 5: (a) Angle of minimum remanent coercivity and the corresponding normalized value of the minimum as a function of CoCrPt-Ox. film thickness. A decrease in the minimum angle indicates the onset of incoherent switching in the material. (b) Angle of minimum

switching field as a function of remanence ratio (squareness) for the thickness series of CoCrPt-Ox. Dashed lines are a guide to the eye.

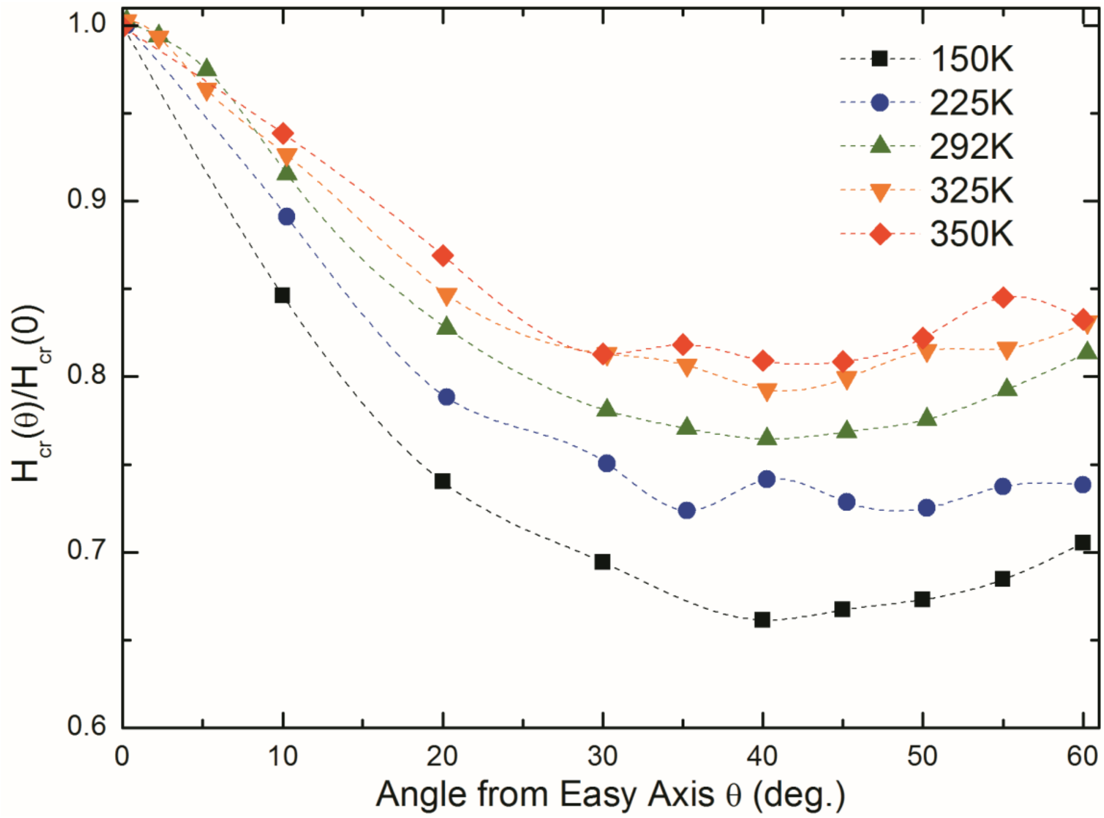

Figure 6: Remanent coercivity H_{cr} as a function of angle for a 6nm thick CoCrPt-Ox. film at five temperatures between 150K and 350K. The angle of the minimum decreases due to increased relative contribution of exchange coupled grains to the reversal process. Dashed lines are a guide to the eye.

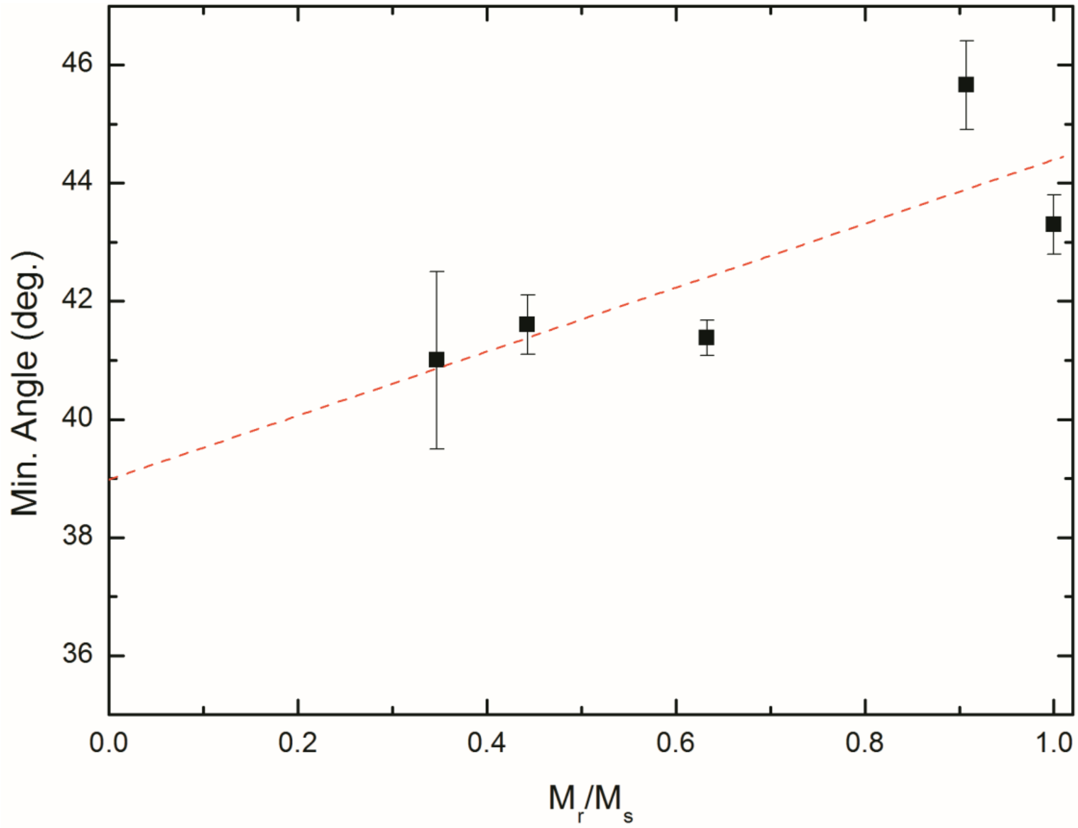

Figure 7: Minimum switching field angle as a function of squareness for a 6nm thick film of CoCrPt-Ox. at five temperature values. Dashed line is a linear fit to the data.

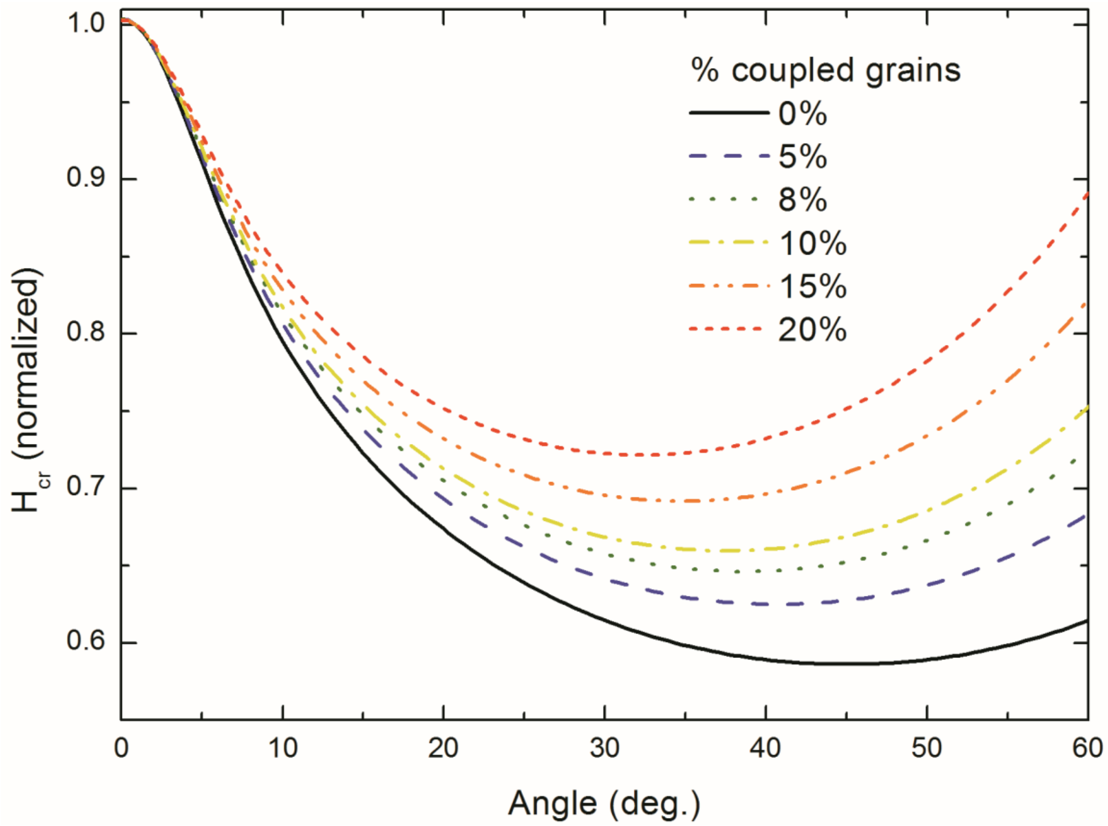

Figure 8: Theoretical reversal curves plotted using equation 1 for six values of the inter-granular coupling fraction *x*.